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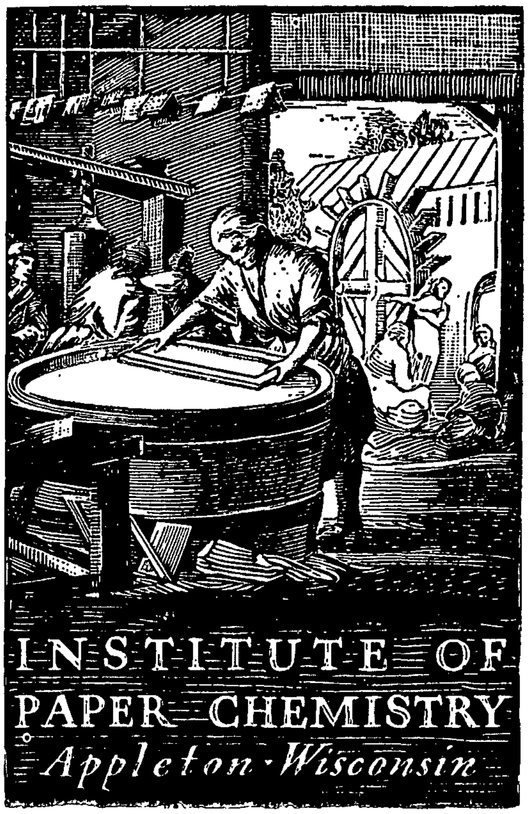

DEVELOPMENT OF COLD-SET ADHESIVE

Project 2696-11

Report One

A Progress Report

to

FOURDRINIER KRAFT BOARD INSTITUTE, INC.

November 7, 1973

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THE INSTITUTE OF PAPER CHEMISTRY

Appleton, Wisconsin

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SUMMARY

A study has been made of cold-set starch corrugating adhesives comprised of pearl cornstarch, ammonium persulfate, a salt such as sodium bisulfite or sulfite, and an alkaline material. Ammonium persulfate modifies the starch to permit the use of higher adhesive solids contents and the bisulfite or sulfite was used to increase the gelation temperature of the adhesive to temperatures as high as 77°C. (171°F.). Alkaline agents such as caustic soda, sodium bicarbonate, and sodium carbonate were added for pH control. The adhesive was prepared by passing a slurry of the starch and chemicals through a jet cooker at temperatures ranging from 230 to 280°F. The solids content of the cooked adhesives ranged from approximately 20-35%. When applied to board in the corrugating operation, the adhesive sets rapidly to a gel as it cools thereby forming a bond between the liner and medium.

The experimental program was divided into two parts. The first part was exploratory in nature and was intended to establish the feasibility of the jet-cooked cold-set adhesive concept. The second part extended these studies in efforts to optimize the conditions for practical corrugating.

Under Part One, an examination was made of several process variables on the starch adhesive properties. Increasing the bisulfite content of the starch slurry at constant persulfate level increased the viscosity and gelation temperature of the jet-cooked product whereas the post addition of sodium sulfite or caustic soda to the cooked adhesive had little effect on viscosity and gelation temperature. The postaddition of borax to a cooked adhesive containing

persulfate and sulfite tended to reduce the gelation temperature. Reheating the gelled adhesive produced a material of greatly increased viscosity compared to the original cooked adhesive thereby making the reheated product unsuitable for corrugating.

The cold-set starch was subsequently utilized as the corrugating adhesive for a standard medium and liner combination under several conditions of persulfate: sulfite ratio, solids content, pH, and corrugator operating temperatures, speeds, and clearances. Within the limits of the experiments, the persulfate:sulfite ratio and pH had little effect on pin adhesion. Heating the corrugator pressure roll tended to reduce pin adhesion at the lowest corrugating speed. Increasing the pressure on the pressure roll appeared to have had a beneficial effect on adhesion but the results were too limited to be conclusive. While the cold-set adhesive was found to provide acceptable "green" bond in most cases, the "dry" bond tended to be inadequate due largely to cohesive failure. However, the pin adhesion values in several cases approached those of the reference two-step adhesive and the program was extended on this basis.

In the second part of the program, consideration was given to the effects of adhesive solids content on bonding strength under several conditions of viscosity and gelation temperature. In as much as chemical modification of starch tends to reduce the unit bond strength, it was reasoned that increased adhesive solids would be needed to improve bonding. After examining further the process variables related to viscosity and gelation temperature, a series of corrugator trials was conducted in which the starch adhesive solids content ranged from 22 to 33%; the viscosity from 70 to 420 Brabender units; and the gelation temperature from 59 to 70°C. (138-158°F.). The corrugator was operated at speeds up to 400 f.p.m. under conditions of constant glue roll clearance and temperature but with variation in the pressure roll temperature and pressure. A steam injection-water bath was utilized as a starch-holding tank during this series in an effort to provide better uniformity and to prevent viscosity build-up which is believed to have occurred in the earlier trials. Pin adhesion values comparable to the reference two-step adhesive were attained at a solids level of 23% and the values increased significantly at optimum viscosity as the adhesive solids increased to 33%. Within a given narrow solids range, pin adhesion was lowest at the highest viscosity. The maximum adhesion occurred at intermediate viscosity and gelation temperatures. Hence, the cold-set starch formulation was found to provide adhesion values comparable to the conventional adhesive without the need of resorting to unusually high solids levels. Subsequent to this base-line study, a second series of trials was conducted in which several variations of the cold-set adhesive were tested at approximately 23% solids at corrugating speeds up to 660 f.p.m. Applications were made to wet strength liner and medium as well as the standard components. The wet strength medium tended to fracture at high corrugating speeds but satisfactory runs were made with the standard components. The cold-set adhesive provided rather consistent pin adhesion values ($\cong$ 70-80 lb.) at 23% solids at all corrugating speeds, whereas the adhesion level declined rapidly at speeds in excess of 300-400 f.p.m. in the case of the conventional adhesive (21% solids). While lack of preheater capacity on the hot-melt corrugator probably contributed to failure of the conventional adhesive, some decline is normally experienced with the two-step adhesive at high corrugating speeds. Hence, the cold-set adhesive was indicated to provide an advantage over the conventional adhesive at only slightly higher solids. The results of this program were considered sufficiently encouraging to warrant continued studies with starch cold-set adhesives.

INTRODUCTION

Traditionally, water-based adhesives have been utilized in the production of corrugated board. This has been due, in part, to the state of the art and to the higher cost of the alternatives. These water-based adhesives are able to make the transition from the fluid condition needed for application to the functioning bonding agent through several mechanisms which produce rapid changes in viscosity. In the case of sodium silicate solutions which are often applied in the range of 30-35% solids, the bond is formed through loss of water to the board components. Sodium silicate has many desirable characteristics, among which are increased rigidity, will develop adhesion without heat, uniform, etc. However, its undesirable characteristics such as adhesion to hot-melt surface, sensitivity to high humidity, high alkalinity, lack of water resistance, etc., has limited the use of sodium silicate to 3-5% of the corrugating adhesive market.

Starch-based adhesives have been most successful in replacing silicate and, in particular, one formulation has become the standard of the industry. This formulation consists of raw unmodified cornstarch suspended in a solution of sodium hydroxide, borax, and dispersed starch. The dispersed starch or primary acts to provide adhesion as well as maintain the raw starch particles in suspension and to provide the necessary flow properties needed for smooth application. Two stages are involved in the functional bond with this adhesive. The initial or green bond is formed when sufficient heat penetrates to the adhesive to cause the raw starch to swell and imbibe water, thereby forming a gel of sufficient strength to hold the structure together. This bond is said to form within 0.1 to 10 sec. depending upon heat transfer. The final bond strength depends upon removal of the water from the gel into the fiber structure and out of the board, if possible. Several measures have been taken, with some success, to retain the water in the starch adhesive to strengthen the green and dry bonds. Jet cooking procedures and the "no carrier" formulations are included among these.

The approach of the present program proposes that a thoroughly cooked starch should be a stronger adhesive than a partially cooked starch. Further, if the cooked starch adhesive would gel at a relatively high temperature and if application were made at a temperature above the gel point, then the adhesive should form a green bond upon cooling and a final bond by vapor diffusion away from the bonded area. In order to produce an adhesive with this property, consideration was given to a combination of modifying chemicals which are known to reduce the viscosity of the hot cooked starch and to enhance the set-back of the paste. The term "set-back" refers to the transition from a slowly increasing viscosity as the fluid cools to an abrupt elevation due to increased intermolecular association preceding gel formation. Two treatments are known to increase the set-back of modified starch over that of the unmodified product. These are acid modification of the raw starch granules and persulfate treatment as employed in the thermomechanical conversion process. Incorporation of certain salts such as sodium bisulfite or sodium sulfite along with ammonium persulfate to the starch as it is heated in a jet cooker raises the gelation temperature of the cooked paste. Hence, the result is a cold-set adhesive which should form a gel when brought into contact with the cooler surface of the liner. In this manner, an instantaneous green bond should be formed which would not be dependent upon the transfer of heat through the liner. The composite of liner and medium would be used as a heat sink to solidify the adhesive which would then dry by diffusion of moisture into the board structure. Since the functionality of a cold-set adhesive does not depend upon heat transfer in short time intervals, an adhesive of this type should theoretically provide an advantage at high corrugating speeds and/or with heavy weight liner.

A patent (1) assigned to National Starch and Chemical Company describes starch-based adhesives which form a green bond by cooling. However, the examples given refer to starch dextrins and to adhesive solids contents in the range of 32-60%. The work described herein utilizes less expensive pearl cornstarch at solids contents under 36%.

Project 2696-11 was initiated at The Institute of Paper Chemistry for the purpose of developing a cold-set adhesive for corrugated board using starch as the basic raw material. The cold-set adhesive concept considered herein is similar in application to conventional hot-melt adhesives in that both are applied at an elevated temperature and forms a bond by cooling. On the other hand, it differs in that the components of hot-melts normally are not dispersible in water whereas the starch is.

The study was carried out in two parts. Part One consists of a study of the effect of various starch modifiers on the "adhesive" characteristics of starch. Part Two involves a study of the effect of adhesive solids content on bonding strength under several conditions of viscosity and gelation temperature.

A. PART ONE

EXPERIMENTAL

General Procedures

Starch Slurry Make-up

A commercial pearl cornstarch was dispersed at approximately 24% solids in tap water in the presence of a specified amount of sodium bisulfite ($NaHSO_3$) or sodium sulfite (Na_2SO_3). In a typical batch, the bisulfite or sulfite was dissolved in 4500 ml. of room-temperature tap water in an 8-liter stainless steel beaker. The salt was dissolved by stirring with a Lightnin' mixer. Sodium hydroxide, sodium bicarbonate, or sodium carbonate was then added in a specified amount and dissolved in the same manner. To this solution was added 1500 g. (o.d. basis) of pearl cornstarch followed by thorough stirring. Ammonium persulfate [$(NH_4)_2S_2O_8$] was added last and stirred in for a few minutes. The amounts of persulfate, bisulfite, and sulfite added were, in all cases, expressed in terms of percentages based on dry weight of starch. The pH of the adhesive slurry was recorded before cooking.

Jet Cooking Procedure

The jet cooker utilized on this project was modified during the course of the experimental program. The unit finally used in preparing the cold-set adhesive is shown in Fig. 1. At the start of a run, warm water was fed to the hopper and the steam pressure was adjusted until the desired operating temperature was attained, 230-280°F. When steady state conditions were reached and a low level of water remained in the hopper, the raw starch slurry was fed to the hopper. The first 200-500 ml. of cooked adhesive was discarded and the remainder collected in a preheated Dewar flask or other container having some means of maintaining

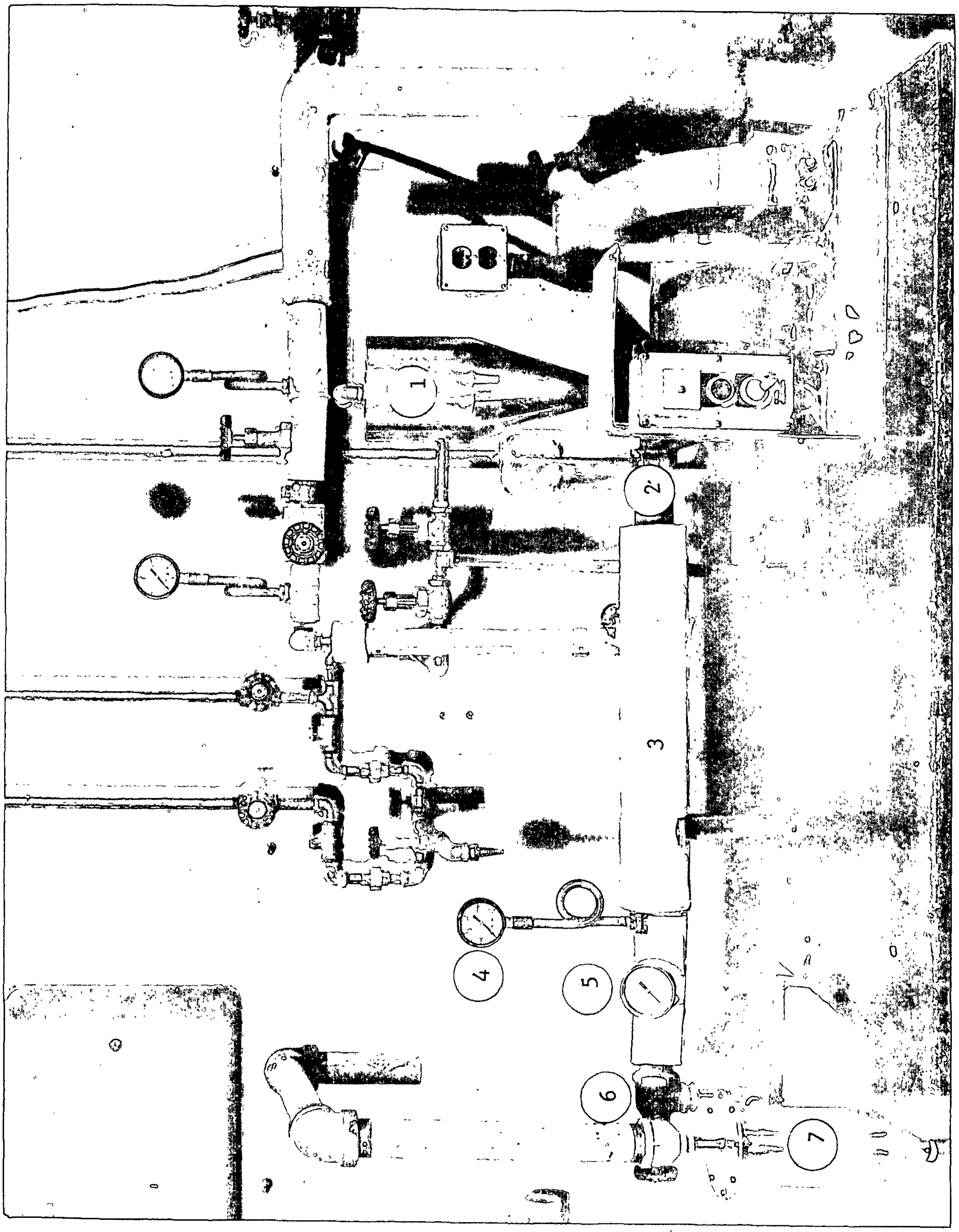

Figure 1. Jet Cooker. 1. Hopper; 2. Moyno Pump; 3. Jet Reaction Chamber; 4. Pressure Indicator; 5. Temperature Indicator; 6. Throttling Valve; 7. Separator

the temperature of the adhesive above the gelation temperature. Initially, large batches of adhesive for corrugator trials were maintained at elevated temperature in a stainless steel vessel wrapped with strip heaters: however, this system was not entirely satisfactory and a steam-heated water bath was utilized in Part Two of the program.

Viscosity, pH, and Solids Determinations

Viscosity and pH measurements were made on the cooked adhesive. The primary instrument used for measuring viscosity was the Brabender Amylograph Viscograph. This device is a recording rotational viscometer having a built-in capability of measuring viscosity as a function of a continually changing temperature, of rotational speed, and of time at constant temperature and rotational speed. The major use of this device in the current program was to determine the temperature at which cooling resulted in a viscosity increase of 500 units. This temperature was arbitrarily referred to as the "set" or "gelation" temperature for purposes of the present program. It is not a generally accepted term. In practice, the cooling cycle was started after the adhesive viscosity had stabilized at 95°C. (203°F.). The viscosity of the hot paste was recorded at 95°C. at rotational speeds of 100 and 190 r.p.m. in most cases. Stein-Hall flow viscosity was measured in a few cases.

Solids determinations were made by taking weighed adhesive samples to dryness at 105°C. These determinations were primarily made on the jet-cooked adhesive and, in some cases, prior to jet cooking.

Corrugating and Testing

The operating conditions of the corrugator were varied in efforts to obtain desired results from the cold-set adhesives. A standard 42-lb. liner and

26-lb. semichemical medium were used in Part One. The adhesive roll clearance in most cases was set at 0.010 or 0.012 inch. The pan temperature varied from 162 to 187°F. and the glue roll from 162 to 190°F. over the course of the corrugator trials. In most trials the pressure roll was either at ambient temperature ($\cong$ 80°F.) or at 330°F. A few trials were run at intermediate pressure roll temperatures. Corrugating speeds ranged from idle to 400 f.p.m. For purposes of control, a single run was made with a conventional two-component starch adhesive under normal operating conditions. The corrugated board was subsequently conditioned at 73°F. and 50% R.H. and tested for pin adhesion.

The Effect of Process Variables on the Starch Adhesive Properties

In order to apply the cold-set concept to practical corrugating operations, it is necessary to control the gelation temperature according to the needs of the corrugating process. In this direction and as a preliminary to subsequent corrugator trials, a series of starch cooks was prepared incorporating 0.5% of ammonium persulfate, 0.3% of sodium hydroxide (based on dry weight of starch), and varying amounts of sodium bisulfite. The procedures employed in preparing and testing the starch were the same as those given in the preceding section. Results are recorded in Table I and the effects of bisulfite addition on viscosity, gel point, and pH are presented graphically in Fig. 2-5.

A second series of starch adhesive preparations was then made to examine the effects of adding sodium sulfite or borax to the jet-cooked starch as a means of controlling the gelation temperature. For this purpose, controls were prepared a) without sulfite and alkali and b) with alkali only. Two adhesive formulations were then prepared incorporating 0.75 and 3.0% of sodium sulfite in the cooked adhesive at approximately 95°C. Ammonium persulfate was added prior to cooking in an amount equivalent to 0.75% based on dry starch. When borax was added in a

TABLE I

THE EFFECT OF BISULFITE ADDITION ON ADHESIVE PROPERTIES

Adhesive No.	Sodium Bisulfite ($NaHSO_3$) Added, % (based on starch)	pH Before Cook	pH After Cook	Solids Content, %	Brabender Viscosity at 95°C., units		Gelation Temp., °C. at 190 r.p.m.
					100 r.p.m.	190 r.p.m.	
1	None	9.5	3.8	22.8	--	140	60 (140°F.)
2	0.125	9.2	5.2	22.0	50	--	59[a] (138°F.)
3	0.250	8.6	5.2	21.5	80	190	62[a] (144°F.)
4	0.375	7.9	4.0	23.0	--	180	62 (144°F.)
5	0.500	7.3	4.3	23.6	--	210	65 (149°F.)
6	0.750	6.9	4.5	22.8	--	310	68 (154°F.)
7	1.000	6.7	5.0	23.0	--	530	75 (167°F.)

[a]Measured at 100 r.p.m.

Note: 0.5% of ammonium persulfate and 0.3% of sodium hydroxide based on starch were added in all cases.

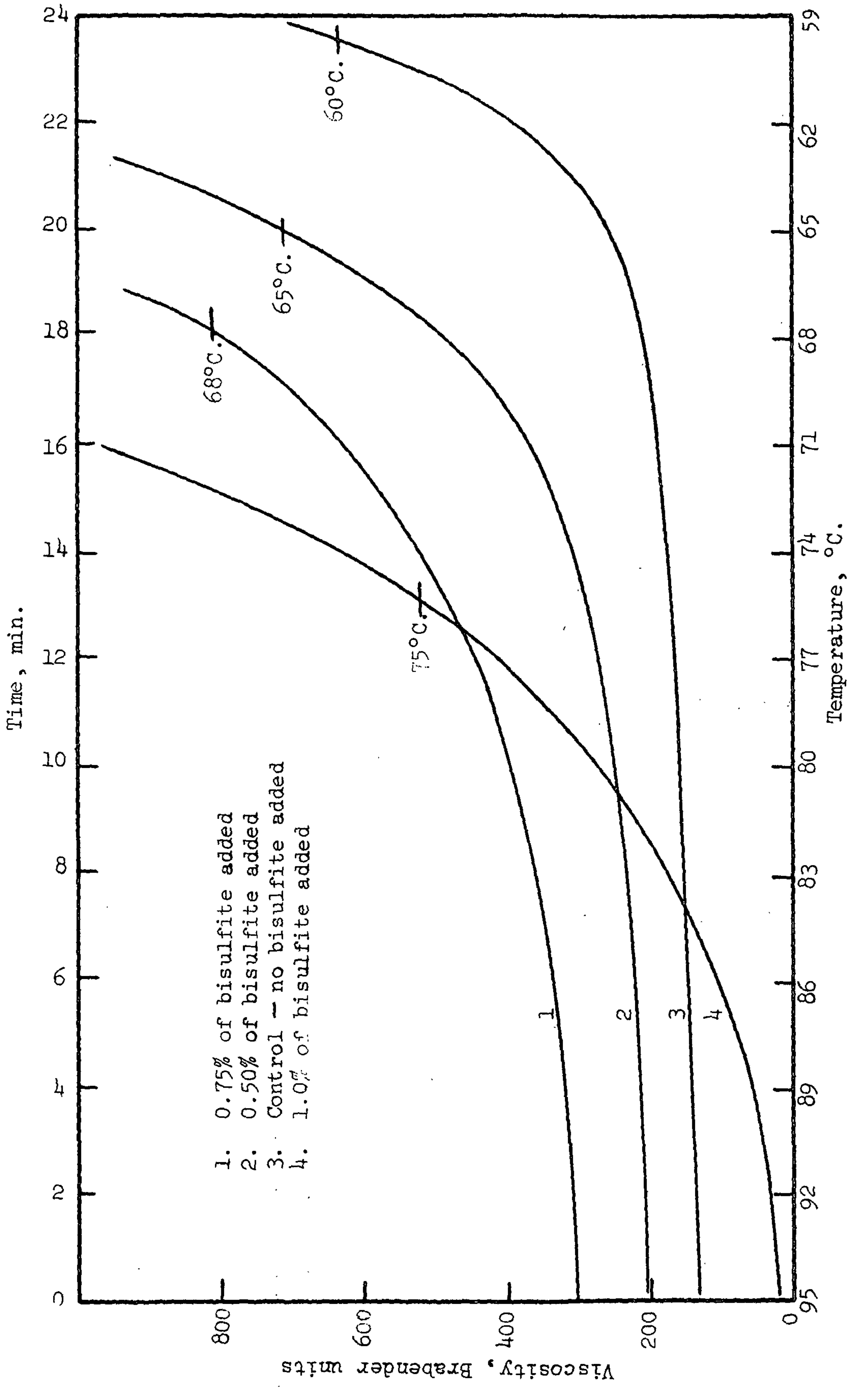

Figure 2. Viscosity vs. Temperature Relationships (190 r.p.m. 0.5% of Persulfate and 0.3% NaOH Added)

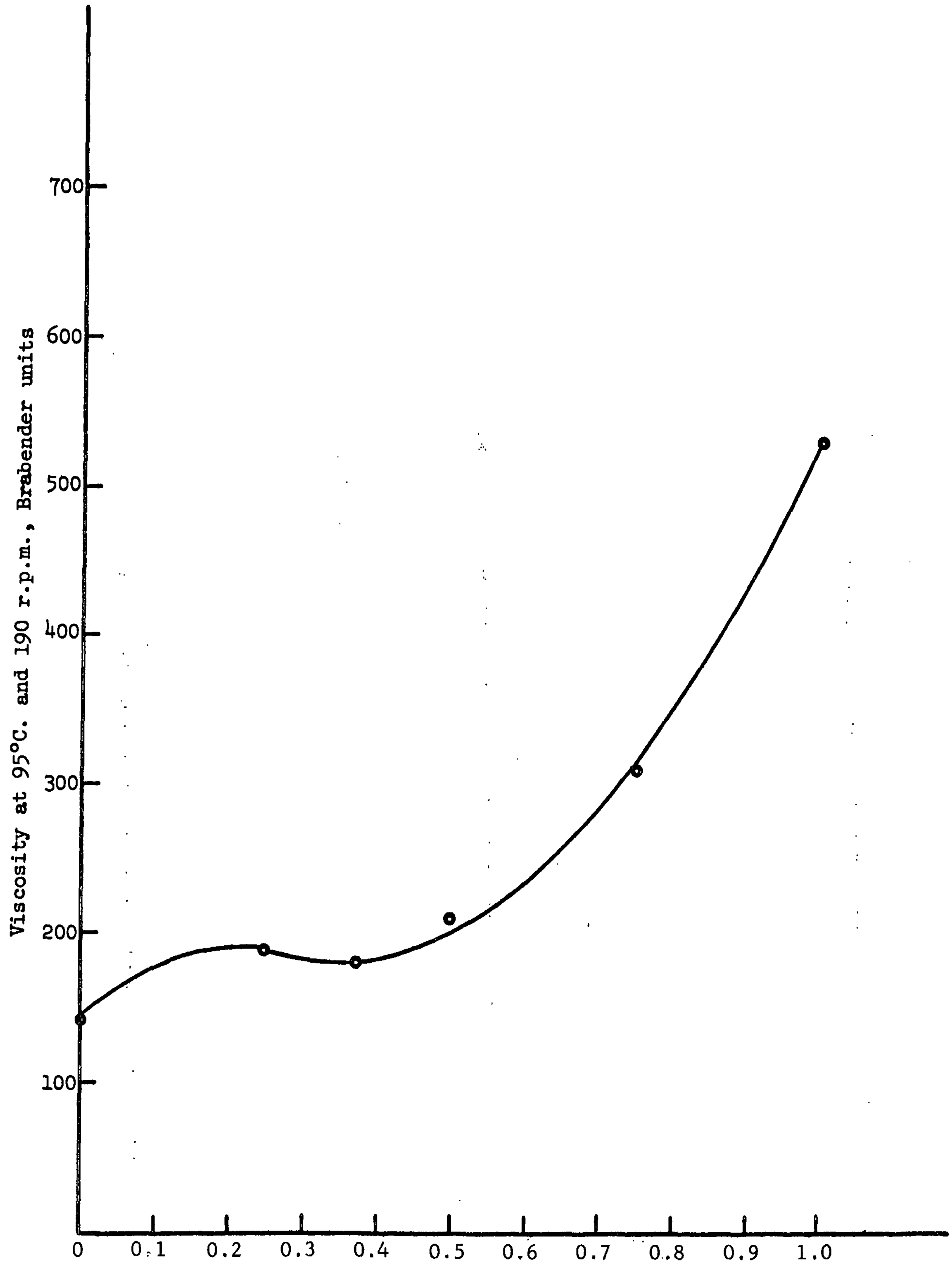

Figure 3. The Effect of Bisulfite Addition on Initial Viscosity (0.5% Persulfate and 0.3% Sodium Hydroxide Added)

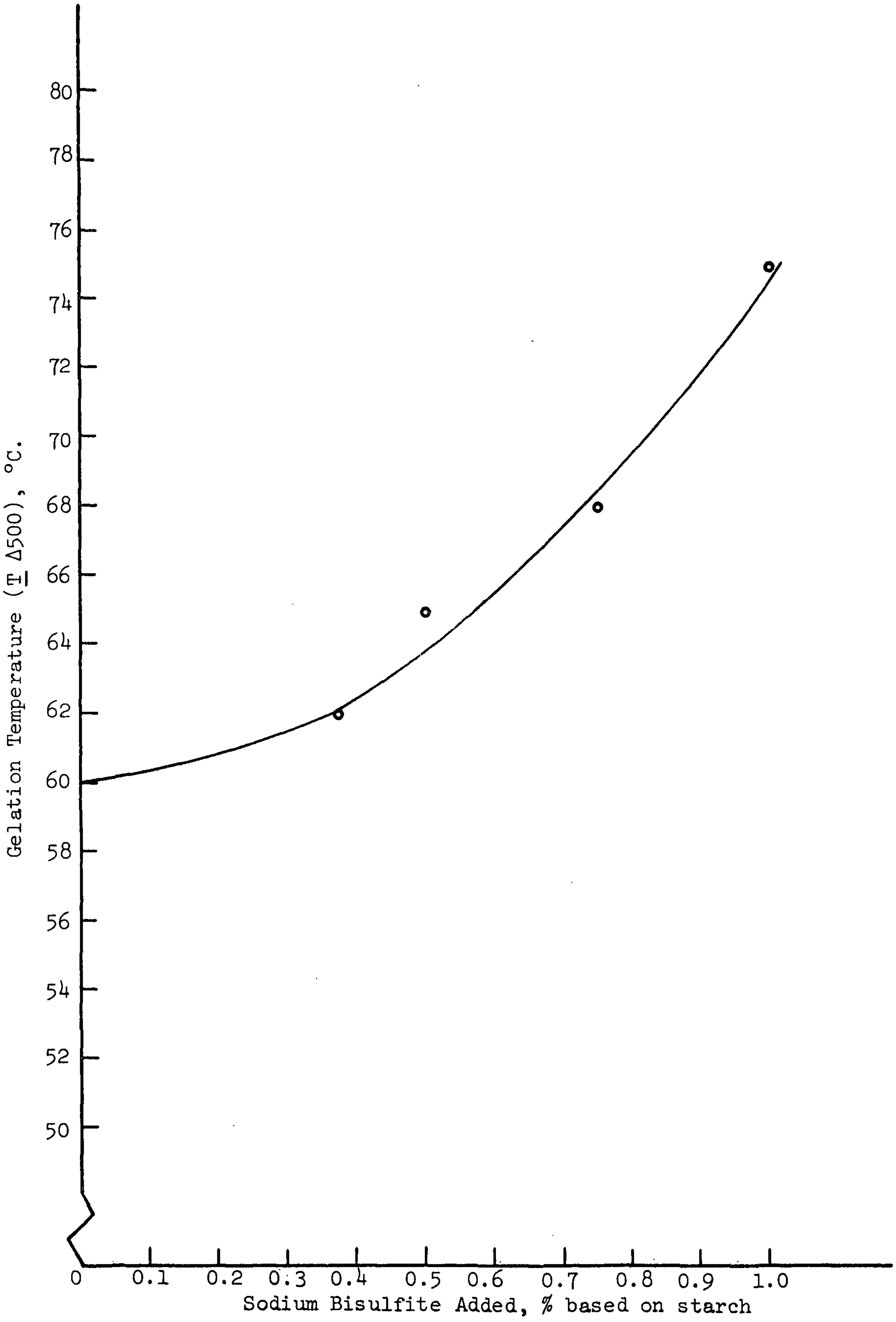

Figure 4. The Effect of Bisulfite Addition on Gelation Temperature
(0.5% Persulfate and 0.3% Sodium Hydroxide Added)

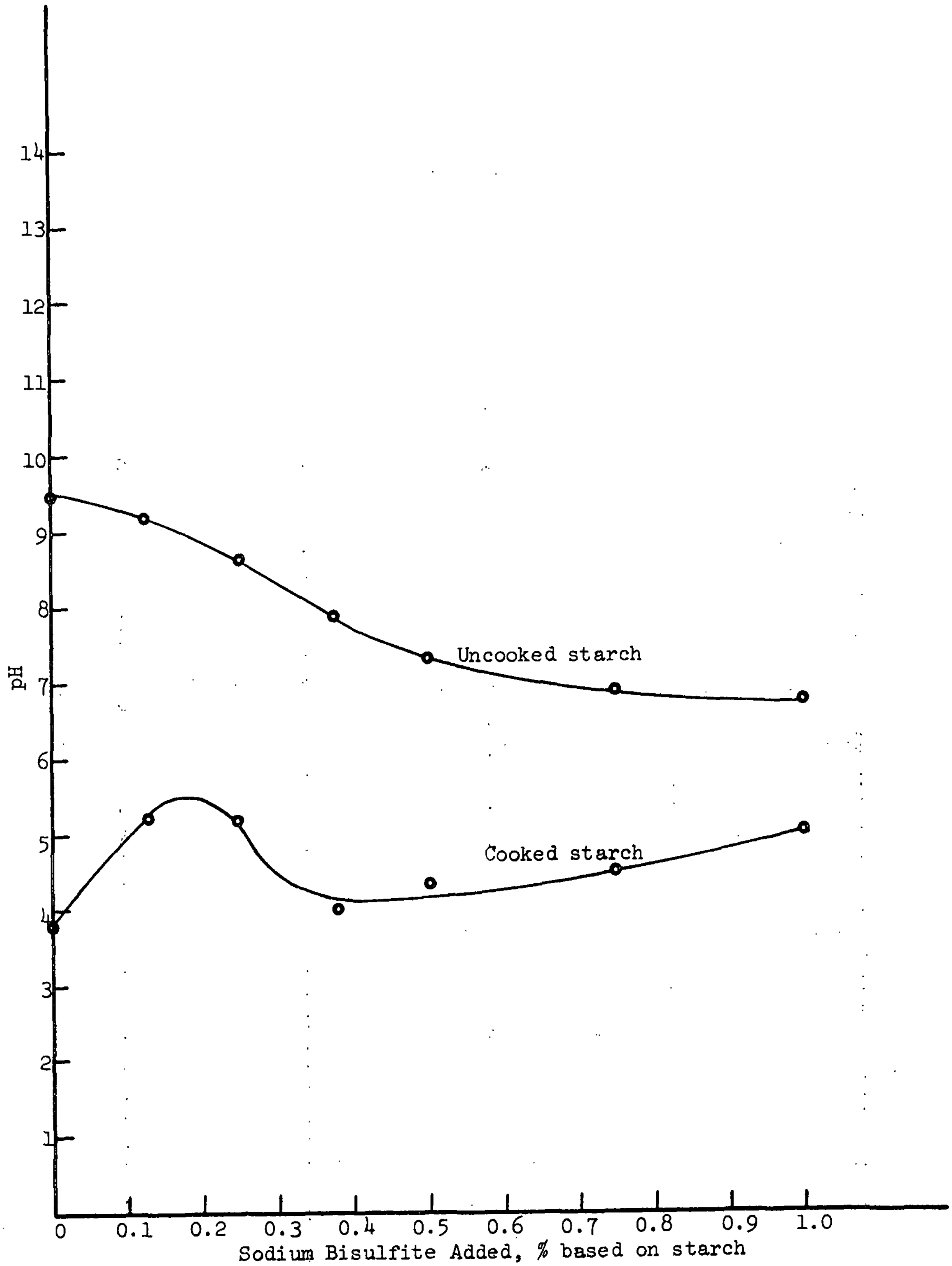

Figure 5. The Effect of Bisulfite Addition on pH (0.5% of Persulfate and 0.3% of Sodium Hydroxide Added)

posttreatment, 0.75% of both persulfate and sodium sulfite was added to the raw starch slurry. In that case, 0.1% of borax (based on dry starch) was added to the cooked adhesive. The jet processing temperature was 230°F. in all cases. The effect of the postaddition of sulfite and borax on viscosity and gelation temperature is shown in Table II.

An examination was also made of the effects of adhesive pH on gelation temperature and viscosity. For this purpose an adhesive was prepared at 230°F. incorporating 0.75% of ammonium persulfate and 0.75% of sodium sulfite. The pH of the cooked starch was varied from 2.3 to 8.5 with increased amounts of sodium hydroxide. Results are recorded in Table III.

In as much as, by design, the processed starch adhesive will tend to gelate rapidly with cooling, accidental chilling would lead to serious problems in corrugating. For this reason, a limited examination was made of the effects on viscosity of reheating the gel. To this end, several adhesives were prepared at 230°F. incorporating 0.5% of persulfate and either 0.5 or 0.6% of sodium sulfite based on dry starch. Sodium hydroxide was added in two cases for pH adjustment. As soon as the adhesive had reached the gelation temperature, i.e., the 500-unit increase in viscosity, the paste was reheated to 95°C. and held at that temperature for 10-15 minutes to stabilize the viscosity reading. The effects of reheating the adhesive on viscosity is shown in Table IV.

Corrugator Trials

On the basis of the information provided in the preliminary work, corrugator trials were made with cold-set adhesives prepared at either 230 or 250°F. incorporating persulfate/sulfite ratios of 0.5/0.5, 0.5/0.6, and 0.75/0.75. In the first series, the corrugator was operated without heat on the pressure roll

TABLE II

THE POSTADDITION OF SODIUM SULFITE AND BORAX

Adhesive No.	Sodium Sulfite Added, % (based on starch)	Borax Added, % (based on starch)	pH of Cooked Adhesive	Solids Content, %	Brabender Viscosity at 95°C., units		Gelation Temp., °C. at 190 r.p.m.
					100 r.p.m.	190 r.p.m.	
8	None	None	1.9	26.7	25	80	--
8-A[a]	None	None	6.3	26.7	10	50	52 (126°F.)
9	0.75	None	7.5	30.5	10	50	51 (124°F.)
10	3.00	None	7.8	34.7	25	90	57 (135°F.)
11	0.75	None	6.7	19.9	--	50	50 (122°F.)
12	0.75	0.10	--	19.9	--	40	41 (106°F.)

Note: 0.75% of ammonium persulfate (based on dry weight of starch) was added in all cases. The jet processing temperature was 230°F.

[a]Sodium hydroxide added.

TABLE III

pH VS. GELATION TEMPERATURE

Adhesive No.	pH of Cooked Adhesive	Solids Content, %	Brabender Viscosity at 90°C., units		Gelation Temp., °C. at 190 r.p.m.
			100 r.p.m.	190 r.p.m.	
13	2.3	26.7	210	380	72 (162°F.)
14	3.9	27.6	240	430	72 (162°F.)
15	7.1	29.0	240	410	72 (162°F.)
16	8.5	29.9	240	400	72 (162°F.)

Note: 0.75% of ammonium persulfate and 0.75% of sodium sulfite (based on weight of starch) was added in all cases. The jet processing temperature was 230°F.

TABLE IV

THE EFFECT OF REHEATING THE GELLED ADHESIVE

Adhesive No.	Sodium Sulfite Added, % (based on starch)	pH of Cooked Adhesive	Solids Content, %	Brabender Viscosity at 95°C., units at 190 r.p.m.		Gelation Temp., °C. at 190 r.p.m.
				Initially	After Reheating	
17	0.6	3.3	22.3	550	1080	72 (162°F.)
17-A[a]	0.6	6.3	--	520	950	70 (158°F.)
18[a]	0.5	7.6	21.6	180	1340	67 (153°F.)

[a]Sodium hydroxide added.

Note: 0.5% of ammonium persulfate (based on weight of starch) was added in all cases. The jet processing temperature was 230°F.

in most cases. Sodium bicarbonate or sodium carbonate was utilized for pH adjustment. A description of the conditions employed and the pin adhesion results are given in Table V.

A second series of trials was run in which the adhesive was jet cooked at 250°F. incorporating a fixed ratio (0.5/0.5) of persulfate and sulfite but with varying amounts of alkali to provide a range in adhesive pH. Variations in pressure roll temperature and pressure were included in the series. A conventional two-component starch corrugating adhesive was included for purposes of reference. Results are recorded in Table VI.

DISCUSSION OF RESULTS

The results in Table I and Fig. 2-5 reveal several trends. Increasing the amount of bisulfite has the effect of increasing the gelation temperature and the initial viscosity at constant persulfate level. If the persulfate level were increased to reduce the initial viscosity, the gelation temperature would be reduced (as will be shown in Part Two) but not necessarily in the same manner as reducing the bisulfite while holding the persulfate constant. Figure 5 shows how the pH of the raw slurry and the adhesive varies with the bisulfite level. Persulfate, by itself, produces acidic products and requires acid-consuming reagents to compensate for the reduction in pH. Caustic soda, soda ash, bicarbonate, and calcium carbonate have been used for this purpose. The slight increase in adhesive pH is consistent with the formation of bisulfite addition products with carbonyl groups produced by the action of the persulfate on the starch. It appears that the postaddition of sodium sulfite to the cooked starch has little effect on gelation temperature until a relatively high level is reached as in Adhesive No. 10 (Table II) and this may be due, in part, to the increased adhesive solids content. Borax (Adhesive

TABLE V

CORRUGATOR TRIALS - SERIES ONE

Corrugator Run No.	Adhesive No.	Ammonium Persulfate Added, % based on starch	Sodium Sulfite Added, % based on starch	Jet Cooking Temp., °F.	Solids, %	pH of Cooked Adhesive	Gelation Temp., °C. at 190 r.p.m.	Operating Temp., °F. Pan	Glue Roll	Press Roll	Speed, f.p.m.	Pin Adhesion, lb.	Major Locus of Failure
1	19	0.5	0.6	250	19.3	6.3	--	175	190	Cold	400	40	At liner-adhesive interface and within the adhesive
2	20	0.75	0.75	250	22.3	7.8	73	175	190	Cold	Idle	83	At liner-adhesive interface
3	21	0.75	0.75	230	26.3	7.6	73	184	185	Cold	Idle 400	87 --[a]	At liner-adhesive interface and within the adhesive
4								165	166	Cold	Idle 400	70 —[a]	Within the adhesive
5								165	166	156	Idle 400	61 --[a]	At medium-adhesive interface
6	22	0.5	0.5	250	24.8	7.6	61	182	180	Cold	Idle 400	78 64	At medium-adhesive interface
7								162	162	Cold	Idle 400	47 --[a]	At medium-adhesive interface
8	23	0.5	0.5	250	28.8	7.5	72	187	187	Cold	Idle 400	80 --[a]	At medium-adhesive interface
9								187	187	180	Idle 400	56 --[a]	At medium-adhesive interface
10								187	187	210	Idle 400	59 --[a]	At medium-adhesive interface

[a]The dried bond was too brittle to test for pin adhesion.

Notes: Sodium carbonate or bicarbonate was used for pH adjustment in this series. The clearance was 0.014 inch in Run 1 and 0.010 inch in all other cases.

TABLE VI

CORRUGATOR TRIALS – SERIES TWO

Corrugator Run No.	Adhesive No.	Ammonium Persulfate Added, % based on starch	Sodium Sulfite Added, % based on starch	Solids, %	Stein-Hall Viscosity, sec.	pH of Cooked Adhesive	Operating Temp., °F. Pan	Operating Temp., °F. Glue Roll	Operating Temp., °F. Press Roll	Speed, f.p.m.	Pin Adhesion, lb.	Major Locus of Failure
11	24	0.5	0.5	--	--	6.8	186	186	Cold	Idle	91	Within liner
										100	91	Within adhesive
										200	82	Within adhesive
										400	77	Within adhesive
12						(increased press roll pressure)				Idle	89	At medium-adhesive interface
										100	84	At medium-adhesive interface and within adhesive
										200	73	At medium-adhesive interface
										400	60	At medium-adhesive interface
13							186	186	330	Idle	25	Within adhesive
										100	72	At medium-adhesive interface
										200	61	At medium-adhesive interface
										400	52	At medium-adhesive interface
14	25	0.5	0.5	23.0	80[a]	8.5	188	190	330	Idle	15	Within adhesive
										100	64	Within adhesive
										200	60	Within adhesive
										300	58	Within adhesive
										400	59	Within adhesive
15	26	0.5	0.5	25.0	119[b]	9.7	187	190	330	Idle	28	Within adhesive
										100	59	Within adhesive
										200	57	Within adhesive
										300	66	At liner-adhesive interface
										400	79	At medium-adhesive interface
16	27	Control (conventional two-step starch adhesive)		21	32[c]	--	95	100	330	Idle	91	At liner-adhesive interface
										100	85	At liner-adhesive interface
										200	85	At liner-adhesive interface
										300	73	At liner-adhesive interface and within adhesive
										400	56[d]	At liner-adhesive interface and within adhesive

[a]At 160°F.
[b]At 170°F.
[c]At 100°F.
[d]Inadequate preheater.

Notes: Sodium carbonate and sodium hydroxide used for pH adjustment in this series. The jet cooking temperature was 250°F. in all cases. The clearance was 0.010 inch in Run No. 11 and 0.012 inch in all other cases.

No. 12) appears to lower the gelation temperature and counteracts the effects of the sulfite ion.

Increasing the alkali level of the cooked adhesive (Table III) is shown to have no effect on gelation temperature and only a modest effect on hot viscosity. Reheating the starch paste immediately after reaching the gelation temperature (Table IV) results in greatly increased viscosity at 95°C. Thus, if initial gelation occurs in commercial operations due to extended holding periods or lack of proper temperature control the adhesive must be discarded as it cannot be made usable merely by reheating.

Several effects are evident in the results of the corrugator trials as presented in Tables V and VI. The pin adhesion values in several cases (Adhesives 24-26) approached or equalled those of the conventional two-component adhesive at roughly equal film thickness. The green bond resulting from the cold-set adhesive was adequate in most cases but the "dry" bond tended to be brittle and little or no adhesive strength was attained at 400 f.p.m. in the first series as indicated in Table V. This may result from the manner in which the adhesive dries, possibly incurring internal stresses which causes the bond to develop fractures. Varying the ratio of persulfate to sulfite seems to have little influence on the pin adhesion result. Varying the pH at constant film thickness and pressure roll temperature as recorded in Table VI also appears to have had little consistent effect. The pressure roll temperature is indicated to have some effect on bond strength depending upon corrugating speed. Heating the pressure roll at the idle speed in the first series of tests (Table V) appears to have lowered bond strength although the locus of failure remained the same, i.e., medium-adhesive interface. It will be noted that heating the pressure roll at idle speed in the second series drastically reduced the pin adhesion value whereas the differences were less pronounced at the higher speeds (see Adhesive No. 24). The predominant locus of failure with the cold-set adhesives in the second series was within the adhesive which may again reflect the brittle condition of the bond as previously mentioned. Hence, it appeared that bond strength at low adhesive solids (20-26%) was inadequate. Efforts were accordingly made in Part Two of the program to increase adhesive solids and bond strength.

B. PART TWO

As previously mentioned, Part Two is concerned with a study of the effect of adhesive solids content on bonding strength under several conditions of viscosity and gelation temperature.

EXPERIMENTAL

General Procedures

The procedures utilized in preparing the cold-set starch adhesive and in measuring its viscosity and gel point were similar to those given in Part One with perhaps a few minor changes or variations. Sodium sulfite and sodium hydroxide were the only agents used for controlling gelation temperature and pH, respectively. The pH of the cooked adhesive was adjusted to near neutrality in all cases. The supply of adhesive for each corrugator trial was stored in a covered stainless steel container which, in turn, was stationed in a water bath heated to approximately 85°C. (185°F.) by steam injection. Localized cooling was avoided and improved uniformity was attained in this manner.

The Institute's hot-melt corrugator was again utilized to apply the cold-set adhesive. The standard 26-lb. medium and 42-lb. liner were used in most cases and wet strength components were utilized on a limited basis. The drive mechanism on the corrugator was modified during the course of this work in order to reach speeds in excess of 400 f.p.m. The standard medium and liner could be handled satisfactorily at speeds up to 660 f.p.m. but the wet strength corrugating medium (30-lb.) fractured at speeds in excess of 400 f.p.m. The adhesive film clearance was 12 mils in all cases and the pan roll temperature fell in the range of 181 to 198°F. The pressure roll temperature was either ambient or 310-330°F. and the pressure was arbitrarily adjusted to high or low. The conventional two-step starch adhesive was used as a reference on both the regular and wet-strength components.

Further Examination of Process Variables Related to Starch Adhesive Properties

The results in Part One indicated rather poor cohesive strength in the cold-set adhesive at higher corrugating speeds. It was reasoned that adhesive strength would be improved by increasing the solids at the interface since, in effect, persulfate and other starch modifications tend to reduce the unit bond strength. Hence, the first series of tests in the second part of the program was directed at means of increasing the adhesive solids content within a given range in viscosity and gelation temperature. More specifically, means were sought to produce starch adhesives having:

1. essentially constant viscosity but increasing solids content, and
2. essentially constant solids and gelation temperature but varying viscosity.

In pursuing this work, starch adhesives were prepared at solids contents of approximately 23-24% and 30-35%. In each case changes in viscosity was accomplished by varying the persulfate content or the persulfate/sulfite ratio. Higher cooking temperatures were explored at approximately 24 and 35% solids. The results of these exploratory tests are listed in Tables VII and VIII. The effect of persulfate level on viscosity and gel point at low solids is shown graphically in Fig. 6 and 7. Viscosity and gel point as a function of adhesive solids content are shown in Fig. 8 and 9.

Corrugator Trials

Trials were subsequently run on the hot-melt corrugator at speeds up to 400 f.p.m. utilizing the cold-set adhesive and regular liner and medium. The adhesive solids content in these trials ranged from 22.2 to 33.4%; the viscosity from 70 to 430 Brabender units; and the gelation temperature from 59 to 70°C. (138-158°F.). For each adhesive, the pressure roll was utilized

TABLE VII

THE EFFECT OF PERSULFATE LEVEL ON ADHESIVE PROPERTIES

Adhesive No.	Sodium Sulfite Added, % based on starch	Ammonium Persulfate Added, % based on starch	Cooking Temp., °F.	Solids Content, %	pH Before Cook	pH After Cook	Adjusted pH	Brabender Viscosity, units at 95°C. 100 r.p.m.	Brabender Viscosity, units at 95°C. 190 r.p.m.	Gelation Temp., °C. at 190 r.p.m.
28	0.5	0.25	230	23.0	10.2	9.1	7.0	260	420	75
29		0.50	230	23.6[a]	9.6	7.8	7.0	95[a]	185[a]	64[a]
30		0.75	230	23.4	9.4	7.3	7.0	55[a]	105[a]	61.5[a]
31		1.00	230	23.5[a]	9.2	4.9	7.0	35[a]	60[a]	56.5[a]
32		2.00	230	23.6	8.9	2.9	7.0	25	45	40
33	1.0	0.25	230	23.6	9.9	8.5	7.0	450	760	77
34		0.50	230	24.1	9.6	8.0	7.0	165	300	70
35		0.75	230	23.4	9.6	7.6	7.0	95	180	66
36		1.0	230	23.0	9.1	5.9	7.0	35	80	59
37		2.0	230	23.4	8.8	2.1	7.0	25	45	44
38	0.5	1.0	275	24.2	9.3	3.5	7.0	15	50	50

[a]Average values from two cooks.

Note: 0.3% of sodium hydroxide was added before cooking; the pH of the cooked adhesive was finally adjusted to 7.0 before measuring viscosity and gel point.

TABLE VIII

THE EFFECT OF SOLIDS CONTENT ON ADHESIVE PROPERTIES

Adhesive No.	Ammonium Persulfate Added, % based on starch	Cooking Temp., °F.	Solids Content, %	pH Before Cook	pH After Cook	Adjusted pH	Brabender Viscosity, units at 95°C. 100 r.p.m.	Brabender Viscosity, units at 95°C. 190 r.p.m.	Gelation Temp., °C. at 190 r.p.m.
31	1.0	230	23.5[a]	9.2	4.9	7.0	35[a]	60[a]	56.5[a]
39	1.0	230	29.3	9.3	4.8	7.0	85	140	64
40	1.0	230	33.4	9.3	3.9	7.0	125	245	70
41	1.0	230	$\simeq$ 35	Cooker plugged					
42	1.0	230	$\simeq$ 35	Cooker plugged					
43	1.5	230	$\simeq$ 35	Cooker plugged					
44	1.5	270	35.6	9.0	2.9	7.3	100	210	68

[a]Average values from two cooks.

Notes: 0.5% of sodium sulfite and 0.3% of sodium hydroxide were added before cooking in all cases.
The pH of the cooked adhesive was finally adjusted to $\simeq$ 7.0 before measuring viscosity and gel point.

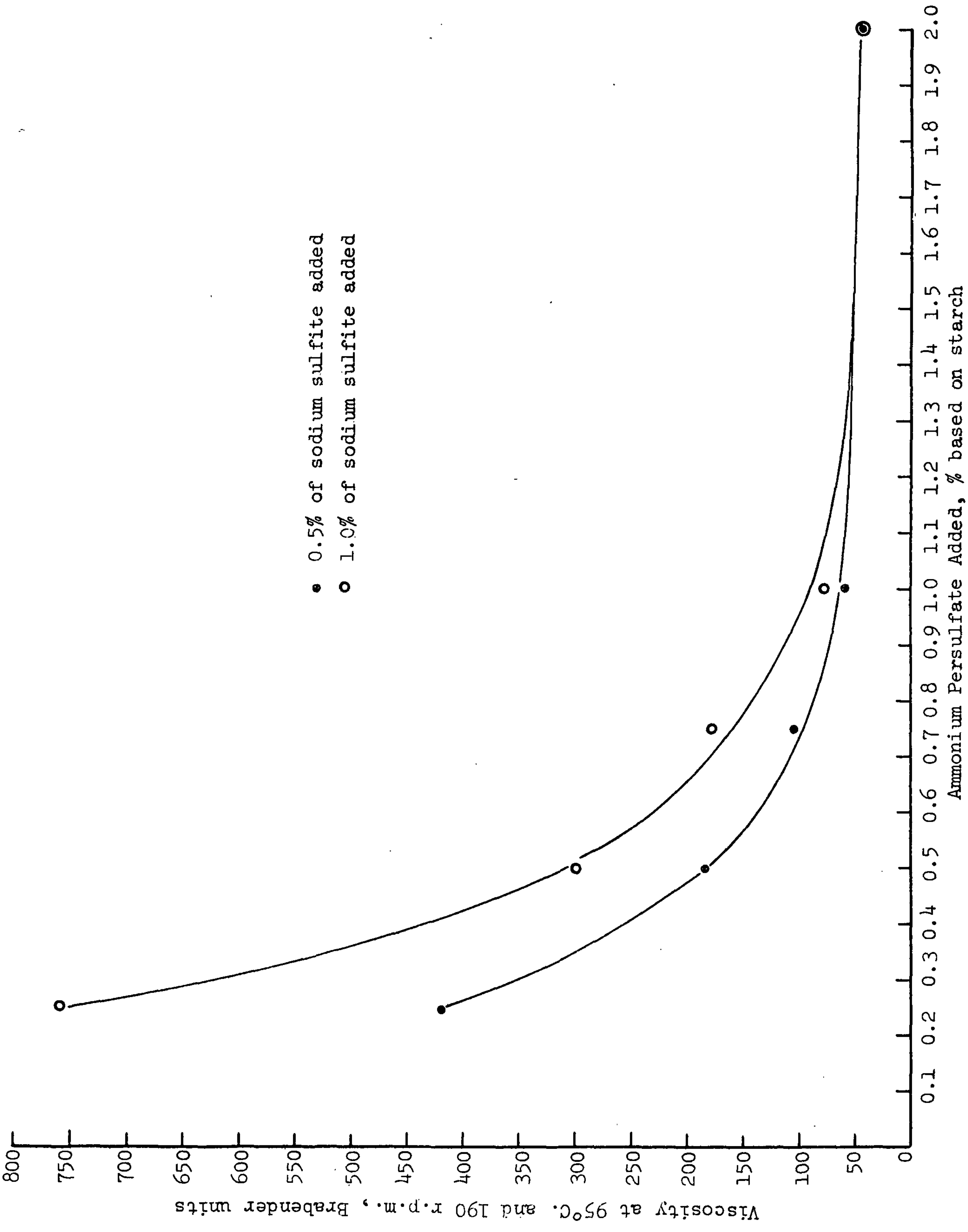

Figure 6. The Effect of Persulfate Level on Viscosity at Constant Adhesive Solids (23-24%)

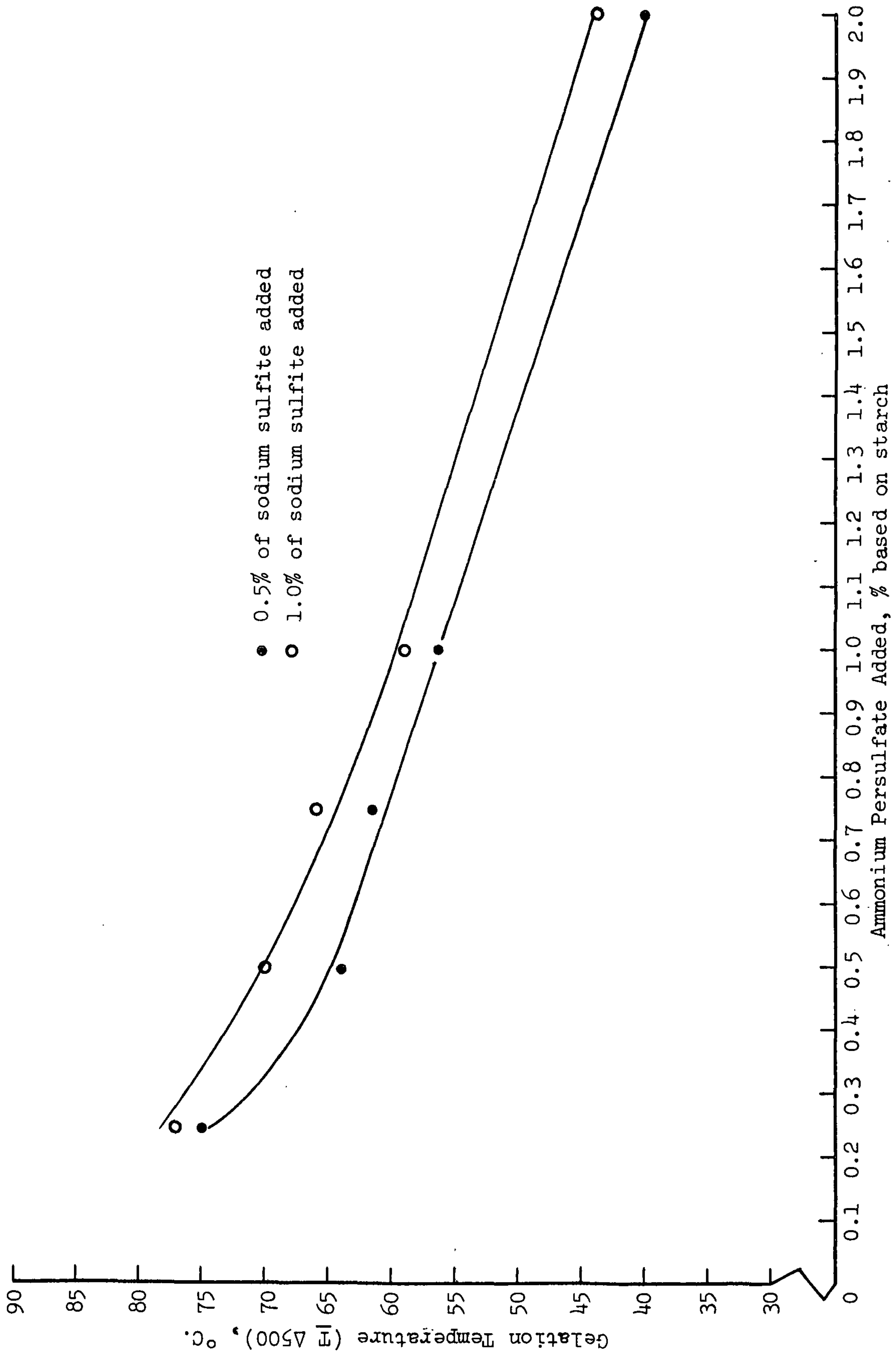

Figure 7. The Effect of Persulfate Level on Gelation Temperature at Constant Adhesive Solids (23-24%)

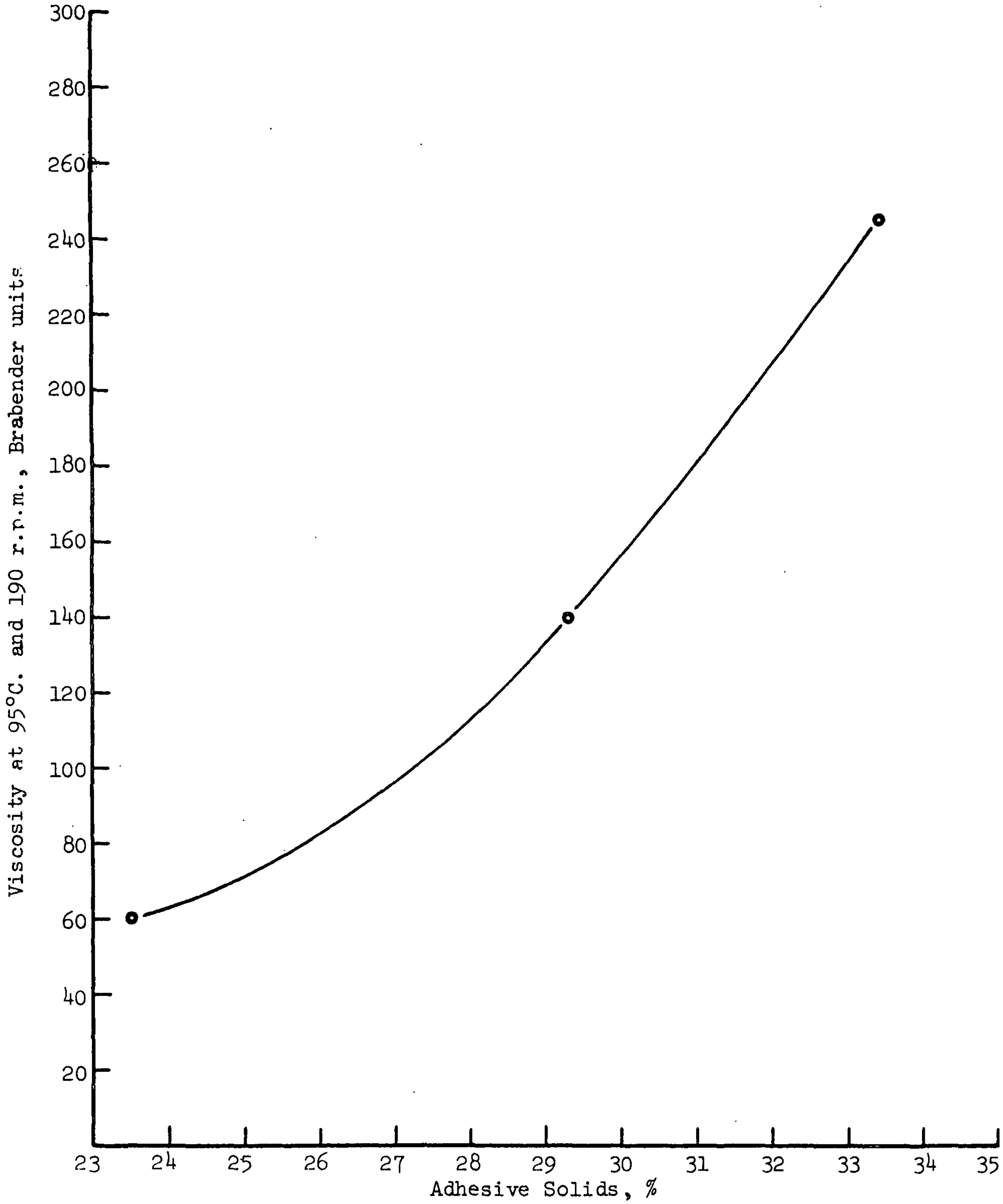

Figure 8. Viscosity as a Function of Adhesive Solids at Constant Persulfate and Sulfite Levels (1.0% Persulfate; 0.5% Sulfite)

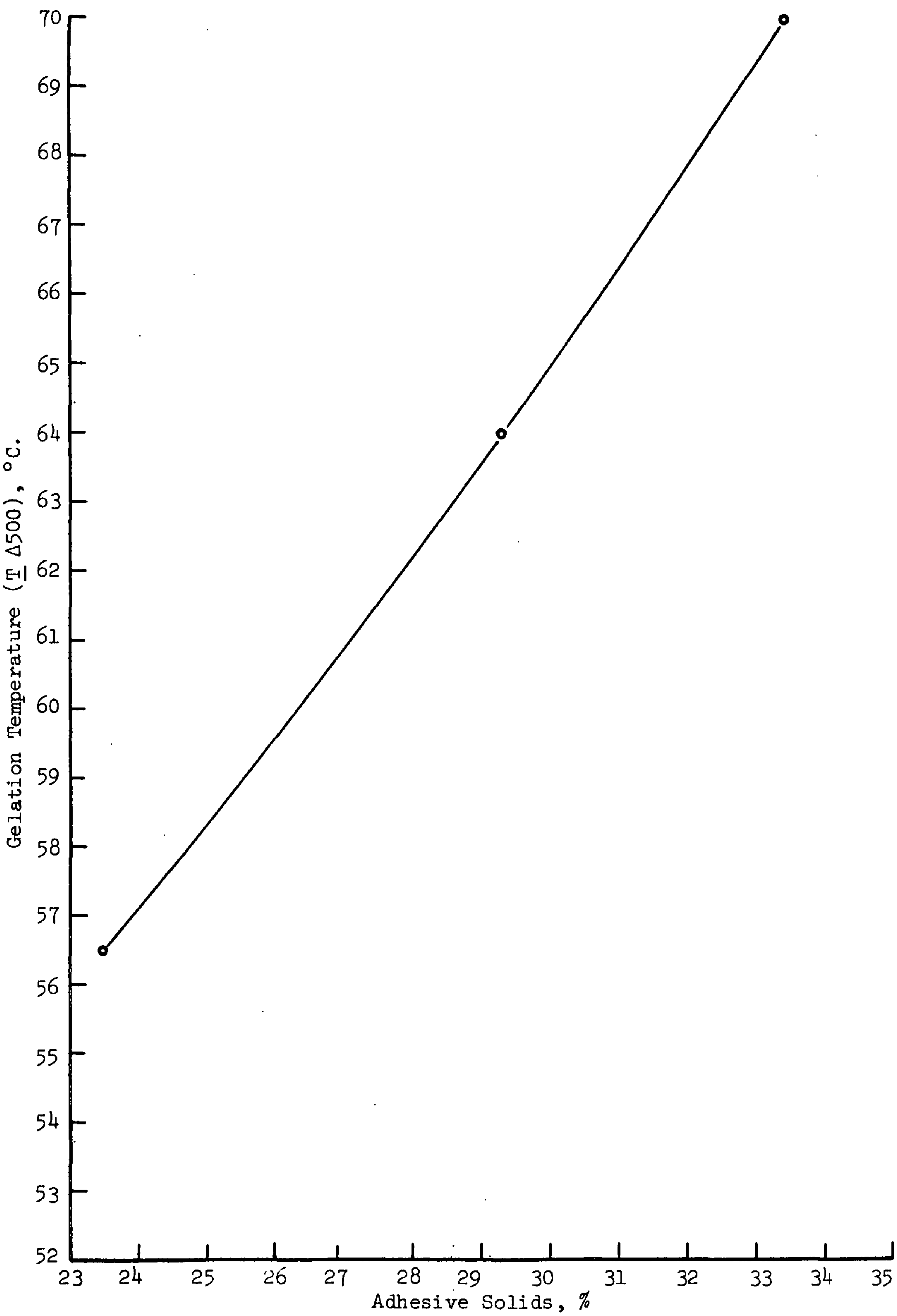

Figure 9. Gelation Temperature as a Function of Adhesive Solids at Constant Persulfate and Sulfite Levels (1.0% Persulfate; 0.5% Sulfite)

at ambient temperature ($\simeq$ 80°F.) and at 310-330°F. In a few cases, the pressure on the pressure roll was adjusted to low and high levels. The pan temperature was maintained at 180-190°F. During the course of these trials it was found that the adhesive tended to increase in viscosity and gelation temperature somewhat after transfer to the heated holding tank. Dilution of the adhesive with hot water was not particularly effective in reducing the viscosity to the original level unless excessive amounts were added. Hence, modest cooling above the gelation temperature produces an increase in viscosity which must be considered in practical corrugating operations.

A conventional two-step corrugating starch was included in this series for purposes of control. The proportions used in the adhesive formulations, together with the pin adhesion values, are given in Table IX. The effect of adhesive viscosity on pin adhesion is shown in Fig. 10. The pin adhesion values used in this case represented the averages obtained at 200 and 400 r.p.m. for all conditions of pressure roll pressure and temperature. Pin adhesion as a function of adhesive solids at optimum viscosity is presented in Fig. 11. The average pin adhesion values at 200 and 400 f.p.m. were again utilized in this case.

Having established a reasonable pin adhesion level at 23% solids and speeds up to 400 f.p.m., the decision was then made to reexamine selected adhesives at higher corrugating speeds since it is here that a cold-set adhesive should have an advantage over the heat-set type. In order to achieve speeds in excess of 400 f.p.m. on the hot-melt corrugator, modification of the drive mechanism was required. Trials were subsequently run at speeds up to 660 f.p.m. utilizing the cold-set adhesive at approximately 23% solids and the conventional starch adhesive at 21% solids. Both regular liner and medium and wet strength components were utilized in these trials. Several variations of the cold-set

TABLE IX

CORRUGATOR TRIALS – SERIES THREE

Corrugator Run No.	Adhesive No.	Ammonium Persulfate Added, % based on starch	Sodium Sulfite Added, % based on starch	Jet Cooking Temp., °F.	Solids Content, %	Final pH	Brabender Viscosity at 95°C.	Gelation Temp., °C. at 190 r.p.m.	Operating Temp., °F. Pan	Operating Temp., °F. Glue Roll	Operating Temp., °F. Press Roll	Relative Pressure on Press Roll	Corrugating Speed, f.p.m.	Pin Adhesion, lb.	Major Locus of Failure
17	45	0.5	0.5	230-240	24.1	8.0	145	63	186	190-195	Cold	Low	Idle	83.0	At medium-adhesive interface
													100	80.2	At medium-adhesive interface
													200	77.4	At medium-adhesive interface
													400	70.6	At medium-adhesive interface
18	46	0.5	0.5	230	23.0	8.0	185	65	190	190-195	Cold	High	Idle	82.8	At medium-adhesive interface
													100	82.8	At medium-adhesive interface
													200	75.2	At medium-adhesive interface
													400	77.8	At medium-adhesive interface
19	46	0.5	0.5	230	23.0	8.0	185	65	182	190-195	Hot[a]	High	Idle	53.0	At medium-adhesive interface
													100	71.4	At medium-adhesive interface
													200	77.8	At medium-adhesive interface
													400	75.8	At medium-adhesive interface
20	47	1.0	0.5	230-240	24.4	6.0	70	59	181	190-195	Cold	High	Idle	86.4	At medium-adhesive interface
													100	71.4	At medium-adhesive interface
													200	75.4	At medium-adhesive interface
													400	70.2	At medium-adhesive interface
21	47	1.0	0.5	230-240	24.4	6.0	70	59	181	190-195	Hot[a]	High	Idle	50.2	At medium-adhesive interface
													100	73.6	At medium-adhesive interface
													200	75.2	At medium-adhesi e interface
													400	71.8	At medium-adhesive interface
22	48	0.5	1.0	240	22.2	7.6	430	69	190	190-195	Cold	High	Idle	76.0	At medium-adhesive interface
													100	73.0	At liner-adhesive interface
													200	67.6	At liner-adhesive interface
													400	67.4	At medium-adhesive interface
23	49	0.5	1.0	240	22.2	7.6	430	69	182	190-195	Hot[a]	High	Idle	36.2	At medium-adhesive interface
													100	61.2	At medium-adhesive interface
													200	63.0	At medium-adhesive interface
													400	68.0	At medium-adhesive interface
24	50	1.0	0.5	240	31.3	7.0	305	69	186	190-195	Cold	High	Idle	96.8	At medium-adhesive interface
													100	81.6	At medium-adhesive interface
													200	72.8	At medium-adhesive interface
													400	69.0	At medium-adhesive interface
25	50	1.0	0.5	240	31.3	7.0	305	69	185	190-195	Hot[a]	High	Idle	64.2	At medium-adhesive interface
													100	78.8	At medium-adhesive interface
													200	72.4	At medium-adhesive interface
													400	75.2	At medium-adhesive interface
26	51	1.0	0.5	240	31.1	7.2	280	70	184	190-195	Cold	High	Idle	89.4	At medium-adhesive interface
													100	87.2	At medium-adhesive interface
													200	76.8	At medium-adhesive interface
													400	80.4	At medium-adhesive interface

See end of table for footnotes.

TABLE IX (Continued)

CORRUGATOR TRIALS – SERIES THREE

Corrugator Run No.	Adhesive No.	Ammonium Persulfate Added, % based on starch	Sodium Sulfite Added, % based on starch	Jet Cooking Temp., °F.	Solids Content, %	Final pH	Brabender Viscosity at 95°C.	Gelation Temp., °C. at 190 r.p.m.	Operating Temp., °F. Pan	Operating Temp., °F. Glue Roll	Operating Temp., °F. Press Roll	Relative Pressure on Press Roll	Corrugating Speed, f.p.m.	Pin Adhesion, lb.	Major Locus of Failure
27	51	1.0	0.5	240	31.1	7.2	280	70	182	190-195	Hot[a]	High	Idle	70.8	At medium-adhesive interface
													100	83.4	At medium-adhesive interface
													200	80.8	At medium-adhesive interface
													400	84.4	At medium-adhesive interface
28	52	1.2	0.5	240	30.5	7.1	180	66	184	190-195	Cold	High	Idle	93.4	At medium-adhesive interface
													100	86.4	At medium-adhesive interface
													200	81.4	At medium-adhesive interface
													400	84.2	At medium-adhesive interface
29	52	1.2	0.5	240	30.5	7.1	180	66	185	190-195	Hot[a]	High	Idle	79.0	At medium-adhesive interface
													100	81.8	At medium-adhesive interface
													200	80.8	At medium-adhesive interface
													400	78.0	At medium-adhesive interface
30	53	1.5	0.5	280	33.4	6.6	240	64	186	190-195	Cold	High	Idle	111.8	Within medium and at medium-adhesive interface
													100	101.0	At medium-adhesive interface
													200	84.8	At medium-adhesive interface
													400	84.0	At medium-adhesive interface
31	53	1.5	0.5	280	33.4	6.6	240	64	186	190-195	Cold	Low	Idle	122.6	At medium-adhesive interface
													100	98.8	At medium-adhesive interface
													200	84.2	At medium-adhesive interface
													400	89.8	At medium-adhesive interface
32	53	1.5	0.5	280	33.4	6.6	240	64	185	190-195	Hot[a]	Low	Idle	99.2	At medium-adhesive interface
													100	102.0	At medium-adhesive interface
													200	98.6	At medium-adhesive interface
													400	96.6	At medium-adhesive interface
33	53	1.5	0.5	280	33.4	6.6	240	64	183	190-195	Hot[a]	High	Idle	109.4	At medium-adhesive interface
													100	101.8	At medium-adhesive interface
													200	94.8	At medium-adhesive interface
													400	88.0	At medium-adhesive interface
34	54	Control (conventional starch corrugating adhesive)			20.2	12.6	--	--	--	--	Hot[a]	High	Idle	85.4	At medium-adhesive interface
													100	70.4	At liner-adhesive interface
													200	71.4	At liner-adhesive interface and within liner
													400	78.8	Within liner
35	54	Control (conventional starch corrugating adhesive)			20.2	12.6	--	--	--	--	Hot[a]	Low	Idle	86.2	At liner-adhesive interface
													100	85.0	Within liner
													200	75.8	Within liner
													400	79.0	At medium-adhesive interface

[a]310-330°F.

Note: The clearance was 0.012 inch in all cases.

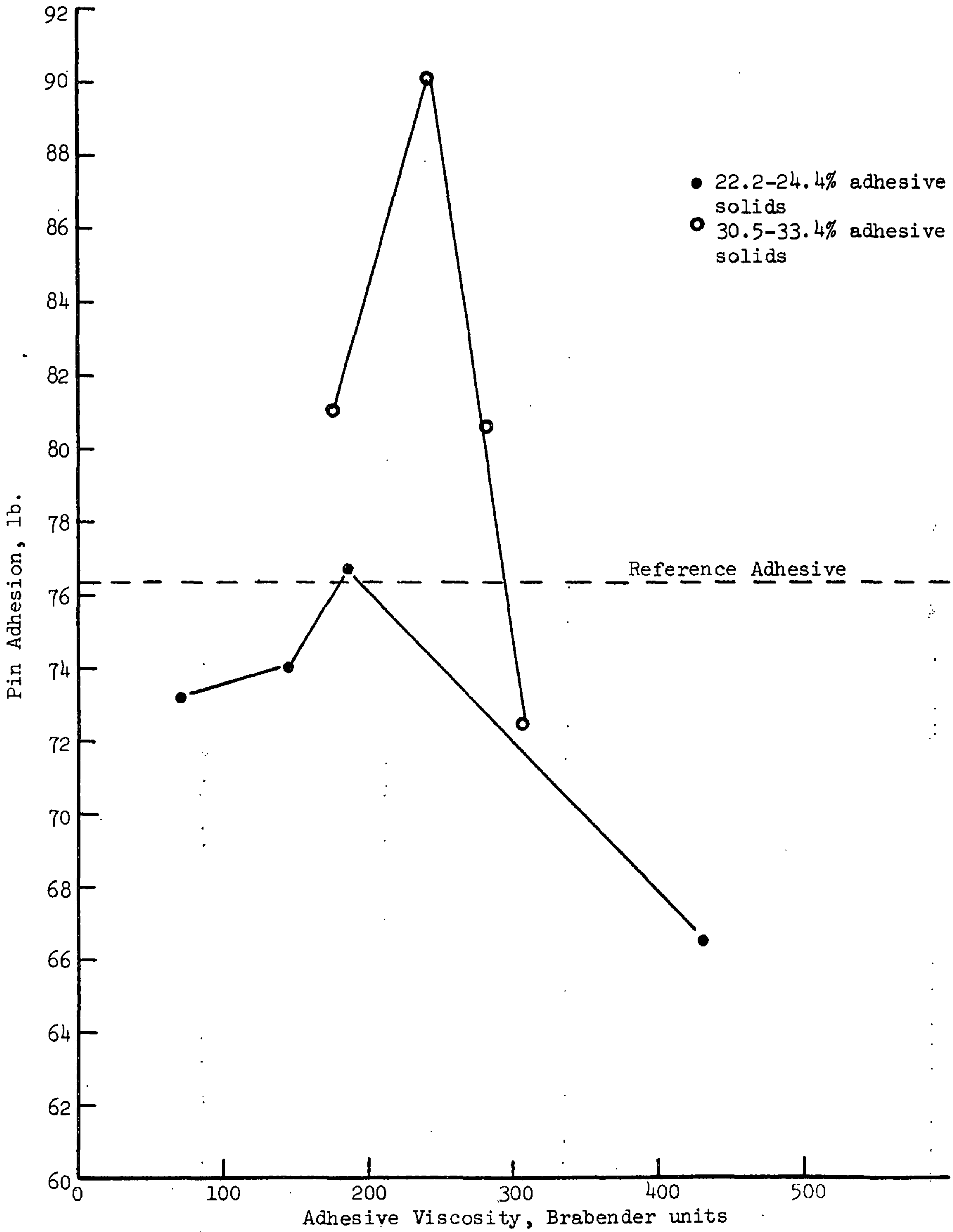

Figure 10. The Effects of Adhesive Viscosity and Solids on Pin Adhesion (Average Pin Adhesion at Corrugating Speeds of 200 and 400 f.p.m.)

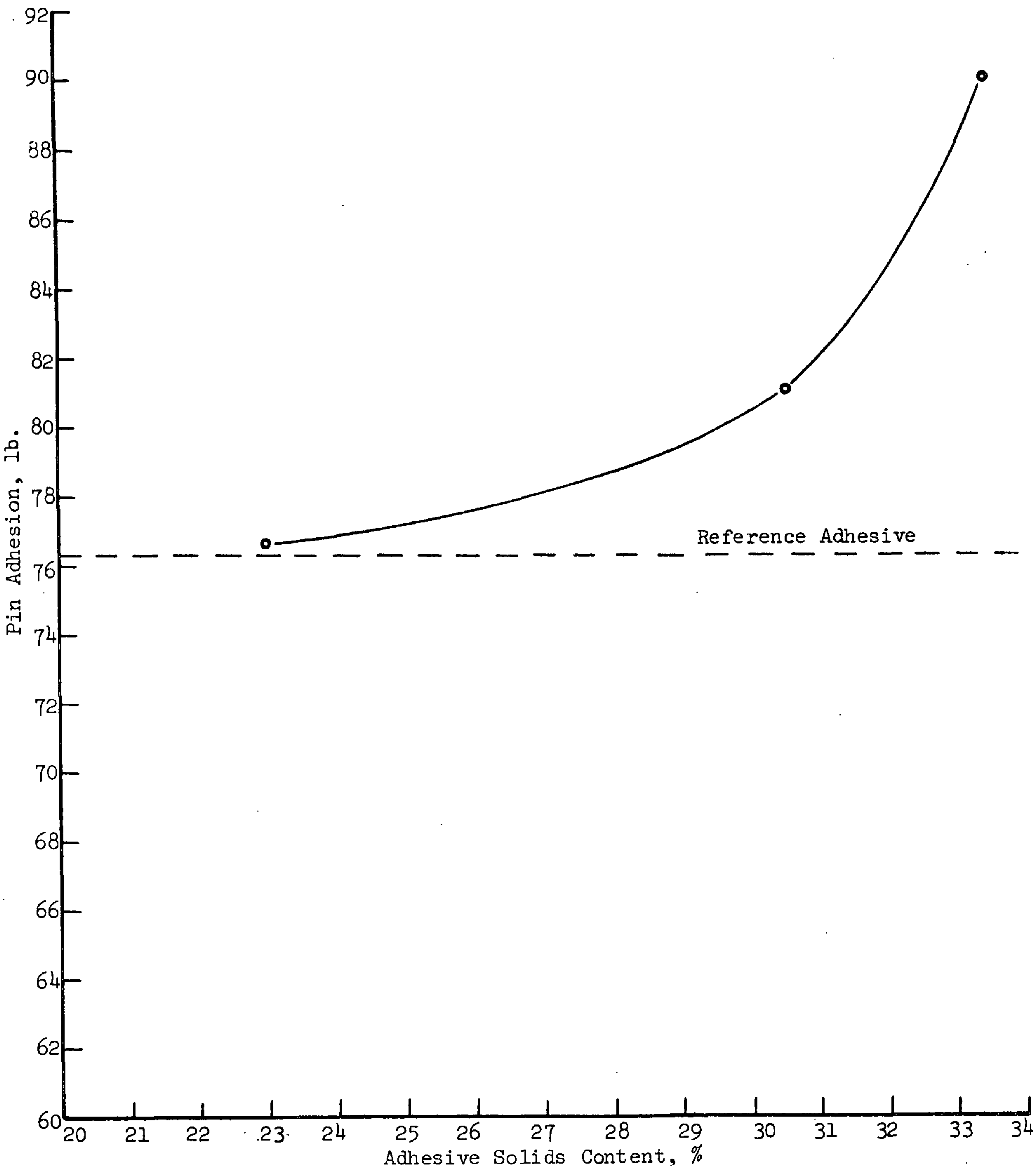

Figure 11. The Effect of Adhesive Solids on Pin Adhesion at Optimum Viscosity (Average Pin Adhesion at Corrugating Speeds of 200 and 400 f.p.m.)

adhesive were included. Two of these varied in viscosity (No. 55 and 56) and the third (No. 57) incorporated 2% of styrene-butadiene (SBR) latex (based on starch) in an effort to reduce an apparent tendency for the persulfate-modified starch to be brittle. The latex was added to the starch after jet cooking. Results are recorded in Table X and pin adhesion as a function of corrugating speed is shown in Fig. 12.

DISCUSSION OF RESULTS

As was expected, increasing the persulfate content at constant sulfite and solids levels reduced viscosity and gelation temperature (Table VII, Fig. 6 and 7). The effect of persulfate was most dramatic at additions less than 1%. Little change in viscosity was indicated at higher persulfate levels and the effect of sodium sulfite was minimized under these conditions. Gelation temperature is shown to follow a more linear relationship with respect to persulfate concentration (Fig. 7) and a rather consistent difference was obtained at the 0.5 and 1.0% sulfite levels. The results in Table VIII indicate that adhesive solids content can be increased to approximately 35% by incorporating 1.5% of persulfate and by increasing the cooking temperature to 270°F. Viscosity tends to increase more rapidly at higher solids, whereas gelation temperature appears to be a linear function of solids content according to Fig. 8 and 9.

The results of this study indicated means of preparing adhesives having 1) roughly comparable viscosity and gelation temperature with varying solids content, and 2) equal solids with varying viscosity and gelation temperature. The latter was considered important as viscosity relates to penetration. This information was utilized in the third series of corrugator trials and the results (Table IX and Fig. 10) show that pin adhesion values comparable to the reference two-step

TABLE X

CORRUGATOR TRIALS – SERIES FOUR

Corrugator Run No.	Liner and Medium	Adhesive No.	Ammonium Persulfate Added, % based on starch	Sodium Sulfite Added, % based on starch	Jet Cooking Temp., °F.	Solids Content, %	Final pH	Brabender Viscosity at 95°C.	Gelation Temp., °C. at 190 r.p.m.	Operating Temp., °F. Pan	Operating Temp., °F. Glue Roll	Corrugating Speed, f.p.m.	Pin Adhesion, lb.	Major Locus of Failure
36	Regular	55	0.5	0.5	230	23.4	7.7	240-270	68	192	190-195	100	75.8	At liner-adhesive interface
												200	83.2	At medium-adhesive interface
												400	70.2	At medium-adhesive interface
												600	69.6	At liner-adhesive interface
												650	67.0	At liner-adhesive interface
37[a]	Regular	55	0.5	0.5	230	23.4	7.7	240-270	68	192	190-195	100	60.8	At medium-adhesive interface
												200	53.2	At medium-adhesive interface
												400	64.6	At medium-adhesive interface
												600	65.4	At medium-adhesive interface
												650	60.0	At medium-adhesive interface
38	Wet strength	55	0.5	0.5	230	23.4	7.7	240-270	68	180	190-195	100	71.6	At medium-adhesive interface
												200	61.2	At medium-adhesive interface
												400	60.6	At medium-adhesive interface
39[b]	Wet strength	55	0.5	0.5	230	23.4	7.7	240-270	68	180	190-195	100	36.6	At medium-adhesive interface
												200	43.4	At medium-adhesive interface
												400	49.4	At medium-adhesive interface
												600	53.4	At medium-adhesive interface
												650	47.6	At medium-adhesive interface
40	Regular	56	0.5	0.5	230	22.6	7.8	160-170	64	198	190-195	100	79.2	At medium-adhesive interface
												200	71.8	At medium-adhesive interface
												400	72.8	At medium-adhesive interface
												600	73.2	At medium-adhesive interface
												660	77.2	At medium-adhesive interface
41	Wet strength	56	0.5	0.5	230	22.6	7.8	160-170	64	198	190-195	100	68.8	At liner-adhesive interface
												200	52.8	At medium-adhesive interface
												400	62.0	At medium-adhesive interface
												600	--[e]	
												650	--[e]	
42[c]	Regular	57	0.5	0.5	230	23.1	7.9	170	65	194	190-195	100	75.4	At medium-adhesive interface
												200	75.4	At medium-adhesive interface
												400	76.2	At medium-adhesive interface
												600	--[e]	
												650	--[e]	
43[c,d]	Regular	57	0.5	0.5	230	23.1	7.9	170	65	194	190-195	100	83.0	At medium-adhesive interface
												200	79.4	At medium-adhesive interface
												400	78.2	At medium-adhesive interface
												600	77.8	At medium-adhesive interface
												650	75.4	At medium-adhesive interface
44	Control (conventional starch corrugating adhesive on regular liner and medium) Adhesive No. 58					21	--	--	--	--	--	100	78.4	Within liner
												200	86.2	Within liner
												300	78.0	Within liner
												400	56.0	At liner-adhesive interface
												500	28.4	Within adhesive
												600	16.8	Within adhesive
												650	18.2	Within adhesive
45	Control (conventional starch corrugating adhesive on wet strength liner and medium) Adhesive No. 58					21	--	--	--	--	--	100	79.8	Within liner
												200	78.4	Within liner
												300	77.6	Within liner
												400	55.6	At liner-adhesive interface
												500	41.6	At medium-adhesive interface

[a] Higher pressure on pressure roll.
[b] Preheater and steam on medium.
[c] 2% of SBR latex (based on starch) added after cooking.
[d] Preheater on medium.
[e] Medium fractured.

Note: The sized medium and liner used in these runs were hydrochem and hydrokraft.

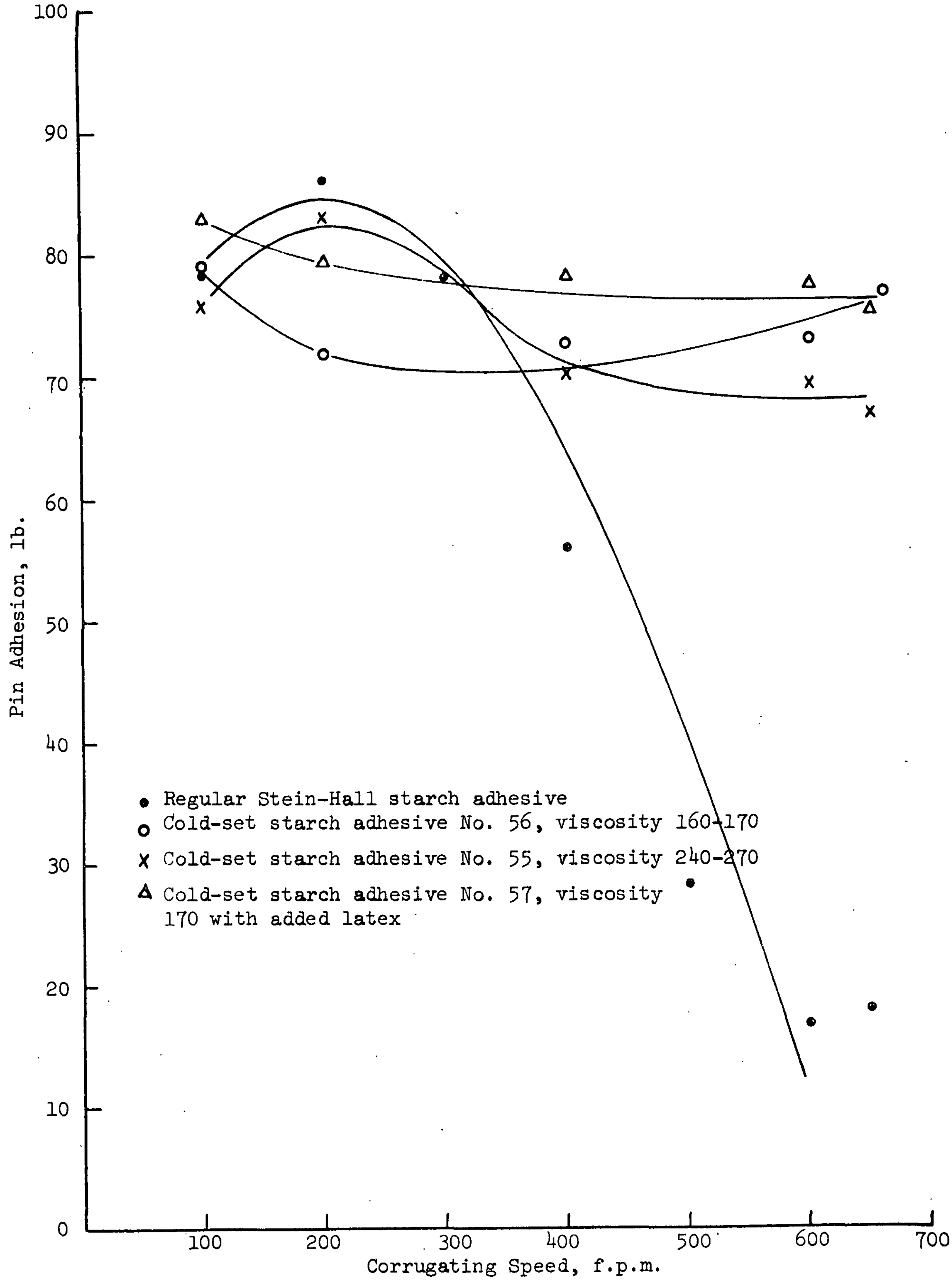

Figure 12. A Comparison of Cold-Set and Regular Starch Corrugating Adhesives at 21-23% Solids (Regular Medium and Liner)

starch adhesive (No. 54) were attained at a solids level of 23% (Adhesive No. 46) and the values increased significantly as the adhesive solids increased to 33% (Adhesive No. 53). Within a given narrow solids range, pin adhesion was lowest at the highest viscosity with the maximum occurring at intermediate viscosity and gelation temperature levels (Fig. 10), although the maximum adhesion, attained at a viscosity of 240 (Adhesive No. 53), also represents the highest solids content adhesive. Small changes in adhesive solids can have a rather pronounced affect on adhesion as shown in Fig. 11 where pin adhesion at optimum viscosity is plotted as a function of solids content. The fact that pin adhesion appears to reach maximum at intermediate viscosity would seem reasonable since excessive penetration may occur at very low viscosity, thereby draining the bonded area of adhesive. Conversely, high viscosity would tend to inhibit penetration leaving a poorly anchored system. High viscosity would appear to be more detrimental to bonding than low viscosity on the basis of the plotted results in Fig. 10.

It may be noted that higher persulfate levels were required in Adhesives 52 and 53 (Table IX) to reach viscosity levels roughly comparable to those of Adhesives 39 and 40 (Table VIII) in spite of the fact that the solids contents were quite similar. These preparations differed only in batch size and in holding time. In the exploratory tests summarized in Tables VII and VIII, small batches were prepared and the adhesive sample for viscosity measurements was taken directly from the jet cooker in a preheated Dewar flask. For purposes of corrugating, larger size batches were required and the adhesive was accordingly transferred from the cooker to the steam-heated holding tank from which the sample for viscosity was drawn. Some cooling occurred in this process resulting in an increase in viscosity. Addition of hot water to the holding tank was not particularly effective in reducing the viscosity unless solids were decreased

3-5%. Hence, increased persulfate levels were required to attain the desired viscosity levels. Conceivably this cooling effect and concomitant increase in viscosity was responsible for some of the difficulties in Part One, where a less efficient holding system was utilized and "dry" bonds were frequently too brittle to test. However, while the "irreversible viscosity" effect may be very significant in laboratory tests, the effect would probably be less critical in commercial operations where the jet cooker could feed the corrugator on a more or less continuous basis.

On the basis of the favorable results obtained at low adhesive solids (Adhesive No. 46) in the third corrugator series, further studies of high solids adhesives were abandoned and a final series of trials was made (Table X) in which the cold-set adhesive was compared to the conventional two-step adhesive at 21-23% solids at corrugating speeds up to 660 f.p.m. Efforts were made to use both regular and wet-strength components in this series; however, the wet-strength medium tended to fracture at higher corrugating speeds and, hence, the data for this system are incomplete. Pin adhesion as a function of corrugating speed for the regular medium and liner is presented in Fig. 12 where it is evident that the cold-set adhesive at $\cong$ 23% solids provided a moderately high and constant level of pin adhesion at all corrugating speeds whereas the values for the conventional adhesive dropped dramatically at corrugating speeds in excess of 300-400 f.p.m. The latter effect was probably magnified by a lack of preheater capacity on the hot-melt corrugator but some decline in adhesion values is normally experienced at higher speeds with the conventional starch adhesive. One problem which persists with the cold-set adhesive is the lack of fiber pull, i.e., failure in the pin adhesion test usually occurred at the medium-adhesive interface which suggests lack of penetration in spite of low adhesive viscosity in several corrugator runs.

This problem would merit consideration in extended studies with the cold-set adhesive.

Hence, the cold-set starch adhesive shows considerable promise at higher corrugating speeds at only slightly higher solids content when compared to the conventional adhesive. The possibility of using it over a range in pH from acidic to alkaline conditions would add versatility to the system with respect to cross-linking to form water resistant bonds. The low concentration of hygroscopic salts in the dry adhesive should also improve the high humidity performance of the bond.

FUTURE WORK

The results obtained in the present program appear sufficiently encouraging to warrant continued work with the cold-set corrugating adhesive concept. Cost considerations would tend to dictate the continued use of starch as the primary component in such systems. Future work would consider means to further reduce adhesive solids content without sacrifice in strength properties. Blends with other adhesives, including polyvinyl alcohol, would be examined for this purpose. Means would be sought to reduce brittleness and enhance plasticity in the persulfate-modified adhesives through incorporation of additives. The potential capacity of the modified starch adhesive to provide water-resistant bonds through cross-linking would also be examined in an extended program.

The development of a practical cold-set adhesive also may have added significance in terms of its compatability with corrugating cold as described in U.S. pat. 3,676,247 issued July 11, 1972 to Andrew W. Morris and Reginald J. Norman and assigned to Australian Paper Manufacturers Limited, Melbourne, Victoria, Australia.

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